

# Competing Ordered Phases in $\text{URu}_2\text{Si}_2$ : Hydrostatic Pressure and Re-substitution

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A persistent kink in the pressure dependence of the “hidden order” (HO) transition temperature of  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  is observed at a critical pressure  $P_c=15$  kbar for  $0 \leq x \leq 0.08$ . In  $\text{URu}_2\text{Si}_2$ , the kink at  $P_c$  is accompanied by the destruction of superconductivity; a change in the magnitude of a spin excitation gap, determined from electrical resistivity measurements; and a complete gapping of a portion of the Fermi surface (FS), inferred from a change in scattering and the competition between the HO state and superconductivity for FS fraction.

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Since its discovery over 20 years ago [1, 2, 3], the moderately heavy fermion compound  $\text{URu}_2\text{Si}_2$  has been the focus of many theoretical and experimental efforts designed to determine the elusive, hidden order parameter associated with the phase transition occurring at  $T_0 \approx 17.5$  K. The transition into this “hidden order” (HO) state is characterized by large anomalies (typical of magnetic ordering) in specific heat, electrical resistivity, thermal conductivity, and magnetization measurements [1, 2, 3, 4, 5, 6, 7]; however, only a small antiferromagnetic moment, insufficient to adequately explain the entropy released during the transition, was detected in low-temperature neutron diffraction experiments [8]. In addition to the puzzling order parameter of the HO state,  $\text{URu}_2\text{Si}_2$  undergoes a transition into an unconventional superconducting (SC) state, which coexists with weak antiferromagnetism (AFM), at  $T_c \approx 1.5$  K. The potential interplay between the two ordered phases of  $\text{URu}_2\text{Si}_2$  as well as the nature of the HO state are underlying problems to our fundamental understanding of the properties of this compound.

In an effort to explain the observed properties of  $\text{URu}_2\text{Si}_2$ , several microscopic models have been proposed [9, 10, 11, 12, 13, 14, 15]. In addition to the theoretical pursuits, many varied experimental techniques have been employed to confirm and/or constrain the proposed models; however, the experimental results fail to converge upon an encompassing microscopic description of the ordered states of  $\text{URu}_2\text{Si}_2$ , but do provide valuable insight when contextually analyzed. Low-temperature neutron diffraction measurements as a function of magnetic field provide evidence that the order parameter of the HO state must break time-reversal symmetry [16], and recent inelastic neutron scattering measurements reveal gapped spin excitations at incommensurate wavevectors [17]. Thermal transport measurements are consistent with the opening of a gap at the Fermi surface (FS), as previously suggested by optical conductivity and specific heat studies [3, 18], depleting carriers and reducing electron-phonon scattering [5, 6]. These exemplary measurements tend to converge upon a description of the HO

state invoking the presence of a FS instability such as a spin density wave (SDW), further suggested by high-field measurements intimating the itinerant nature of the HO state [19].

The application of pressure to  $\text{URu}_2\text{Si}_2$  further convolutes the discussion of the nature of the hidden order parameter as well as the persistence of the SC state. While the HO transition temperature  $T_0$  was seen to increase with applied pressure [4], the SC state was found to be suppressed; however, the reported critical pressures for superconductivity vary greatly from 4-15 kbar, possibly due to disparities in sample quality or high-pressure conditions between experiments [4, 20, 21, 22]. Pressure-dependent neutron scattering, NMR, and  $\mu\text{SR}$  measurements all indicate an abrupt increase in the size of the ordered moment, with the magnitude of the moment saturating to a value consistent with bulk AFM above a currently disputed critical pressure [23, 24, 25, 26]. The interpretation of this increase in moment is currently unresolved, but can be divided into two prevailing descriptions: an inhomogeneous, phase separated scenario where the increase in the moment is due to an increase in volume fraction of the observed, high-pressure moment [25, 26]; and a scenario involving two order parameters describing the staggered magnetization and the HO state, with the details governed by the coupling between order parameters [15, 24].

With the progression of experimental and theoretical investigations into the ordered states of  $\text{URu}_2\text{Si}_2$ , now is a critical time to clarify and categorize experimental observations in order to enhance the understanding of the underlying phenomena. The  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  system provides a unique opportunity to study the effects of pressure on a HO state whose ambient pressure transition temperature is reduced with rhenium concentration  $x$  [27]. In this letter we report new, comprehensive high-pressure electrical resistivity measurements on single crystal samples of the  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  system, emphasizing with greater clarity than previous studies the pressure dependence of the HO state and its relation to the field and pressure dependence of the SC state. A

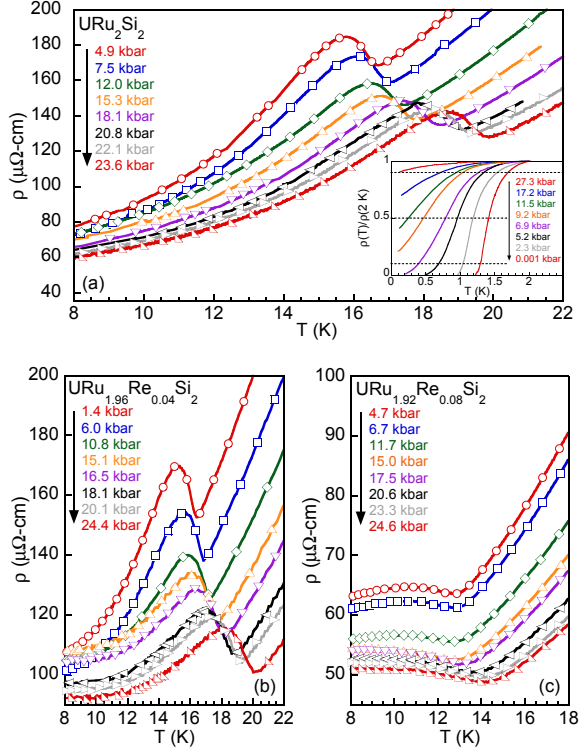


FIG. 1: (color online) (a)  $\rho(T, P)$  of  $\text{URu}_2\text{Si}_2$  as a function of  $T$ . Inset: normalized electrical resistivity,  $\rho(T)/\rho(2\text{ K})$ , vs.  $T$  near  $T_c$  (dashed lines defined in the text).  $\rho(T, P)$  data for specimens of  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  with  $x=0.04$  (b) and  $0.08$  (c).

change is observed in the magnon dispersion gap along with a systematic evolution of the scattering processes in  $\text{URu}_2\text{Si}_2$ . In addition, using a framework developed by Bilbro and McMillan [28], the competition for FS fraction between the HO and SC states is quantified, engendering a consistent depiction of the pressure dependence of the ordered states of  $\text{URu}_2\text{Si}_2$ .

Single crystals of  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  with  $x=0, 0.01, 0.02, 0.04, 0.06$ , and  $0.08$  were grown using the Czochralski method and then annealed at  $900^\circ\text{C}$  for 7 days. The Laue method was used to orient the single crystals, which were subsequently spark cut and polished into electrical resistivity specimens. Electrical resistivity measurements under pressure were performed with a beryllium-copper, piston-cylinder cell using a Teflon capsule filled with a 1:1 mixture of n-pentane:isoamyl alcohol as the pressure-transmitting medium to ensure hydrostatic conditions during pressurization at room temperature. The pressure in the sample chamber was calibrated from the inductively determined, pressure-dependent superconducting critical temperature of a lead manometer.

Displayed in Figure 1(a) are representative electrical resistivity  $\rho(T)$  data illustrating the evolution of  $T_0$ , defined as the local minima occurring between approxi-

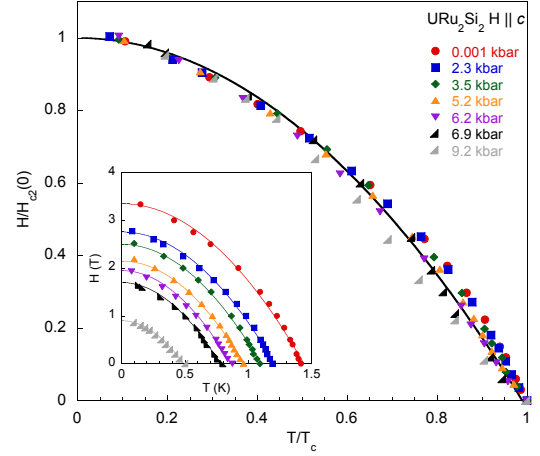


FIG. 2: (color online) Scaled critical field curves,  $H_{c2}(T)/H_{c2}(0)$  vs.  $T/T_c$ , of the SC state of  $\text{URu}_2\text{Si}_2$  for  $P \leq 9.2$  kbar and  $H \parallel c$ . The solid line is a guide to the eye. Inset:  $H_{c2}(T)$  vs  $T$  of the SC state. The solid lines are fits to a parabolic expression yielding values of  $H_{c2}(0)$ .

mately 16.5 K and 20 K, with applied pressure. The data show no qualitative difference in appearance as the characteristic trough and peak structure of the HO transition is preserved to high pressures. The inset of Figure 1(a) shows the electrical resistivity normalized at 2 K,  $\rho(T)/\rho(2\text{ K})$ , emphasizing the pressure dependence of the SC transition. The horizontal dashed lines represent—from top to bottom—90%, 50%, and 10% of the normal state value, with the SC critical temperature  $T_c$  defined as the temperature at which  $\rho(T)/\rho(2\text{ K})=0.5$ . Complete SC transitions were seen up to nearly 7 kbar, and transitions to the 50% value were observed up to approximately 13 kbar; SC fluctuations persisted to the highest pressures measured, although the roles of sample or pressure inhomogeneities are undetermined. Figures 1(b) and (c) display  $\rho(T)$  data near  $T_0$  for specimens of  $\text{URu}_{1.96}\text{Re}_{0.04}\text{Si}_2$  and  $\text{URu}_{1.92}\text{Re}_{0.08}\text{Si}_2$ , respectively. While the absolute value of  $T_0$  and the relative value of the height of the transition in  $\rho$  are altered with  $x$ , the qualitative shape of the feature at  $T_0$  is unchanged, suggesting that, while Re-substitution suppresses  $T_0$ , the physical mechanism responsible for the HO state persists. The  $x$ -dependent changes in the magnitude of the resistivity possibly occur from a change in the relative contribution of impurity scattering with respect to the scattering intrinsic to the HO state.

In addition to the pressure dependence of the SC state seen in the inset of Figure 1(a), field-dependent measurements of the SC state were performed for  $P < 9.2$  kbar with  $H \parallel c$  (inset of Figure 2). The determined critical field curves were fit by a semi-empirical, parabolic expression to extract  $H_{c2}(0)$ , the zero-temperature upper critical field. Using these values of  $H_{c2}(0)$ , the scaled crit-

ical field curves  $H_{c2}(T)/H_{c2}(0)$  were plotted as a function of reduced temperature  $T/T_c$  (Figure 2). The data scale very close to one another using these simple criteria, seemingly indicating that the mechanisms governing the field dependence of the SC state remain unchanged up to the critical pressure.

For all  $x$  measured,  $T_0$  and  $T_c$  are plotted as a function of  $P$  in Figure 3(a), where  $T_0$  evinces a distinct kink in its pressure dependence at 15 kbar, regardless of the ambient pressure value of  $T_0$ . The presence of this kink, when taken within the context of the analysis of Mineev and Zhitomirsky [15], indicates a scenario where there exists no coupling between the order parameters of the HO state and that of the high-pressure AFM phase, and furthermore suggests that sample impurities or uncharacterized strains may be culpable for the observed moment at low pressures. The persistence of this kink and its static position upon reducing  $T_0$  with increased  $x$  is consistent with a vertical or nearly vertical HO/AFM transition occurring at  $P_c=15$  kbar; as this transition is indirectly probed, there can be no unambiguous determination as to the degree of its order. This purported HO/AFM transition has been directly observed in other measurements [21, 24, 29]; however, the reported critical pressures are much lower than 15 kbar, typically near 7 kbar. This discrepancy in  $P_c$  could be due to sample dependence or potential non-hydrostatic conditions present in previous experiments utilizing a mixture of Fluorinert FC70/77, which remains liquid (hydrostatic) only up to approximately 8 kbar [30]. In addition to the persistent kink at 15 kbar,  $T_c$  for  $\text{URu}_2\text{Si}_2$  is suppressed very near  $P_c$ , consistent with previous results suggesting that superconductivity and AFM are mutually exclusive [22]. In fact, if non-hydrostatic conditions are responsible for the early onset of AFM at  $P < 15$  kbar, then one would expect the SC state to be suppressed at these lower pressures as reported by several researchers [20, 21].

It was suggested previously that the HO transition partially gaps a portion of the FS with the remaining ungapped portion undergoing superconductivity at low temperatures [3]. In this scenario, the existence of superconductivity is predicated upon the incomplete FS gap induced by the onset of HO, and, as such, the two ordered states compete for FS fraction. Billbro and McMillan proposed a model to quantitatively analyze the effects of ordered phases competing for FS fraction [28]:

$$T_{c0} = T_c(P)^{n(P)} T_0(P)^{1-n(P)}, \quad (1)$$

where  $T_{c0}$  is the value of the SC critical temperature in the absence of a high-temperature, FS-gapping transition and  $n(P)$  is a measure of the residual ungapped FS—which can be determined from specific heat  $C(T)$  measurements as  $n \equiv \gamma_0/\gamma_{norm}$ , the ratio of the electronic specific heat coefficient above  $T_c$  to that above  $T_0$ . Using the previously determined value of  $n(0)=0.58$  [3], and evaluating Equation 1 at ambient pressure results in a value of

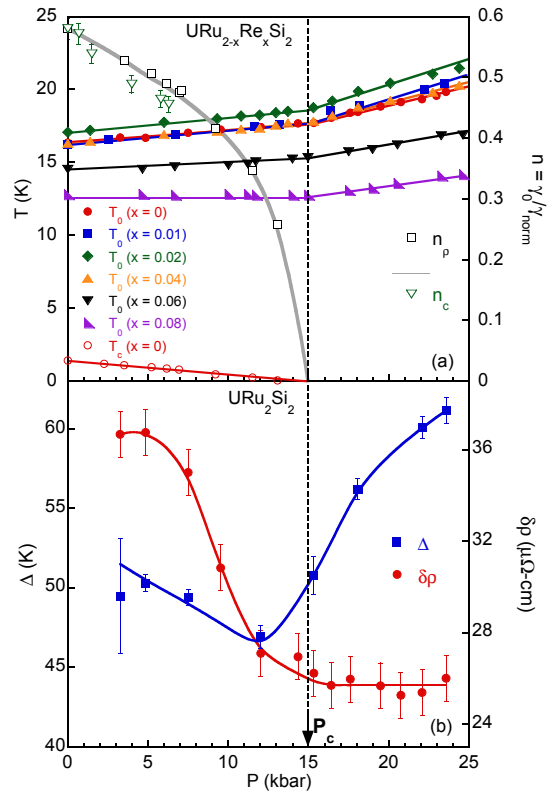


FIG. 3: (color online) (a) T-P phase diagram (left axis) of  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$  showing the evolution of the HO and SC states. An  $x$ -independent kink in  $T_0(P)$  is visible at  $P_c = 15$  kbar. The fraction of the FS left ungapped by the HO transition,  $n \equiv \gamma_0/\gamma_{norm}$  (right axis), vs.  $P$  as determined from  $T_0$  and  $T_c$ ,  $n_p$  (open black squares), and as estimated from previous  $C(T, P)$  measurements [32],  $n_c$  (open, inverted green triangles). (b) Gap in the magnon dispersion  $\Delta$  (left axis) from fits to  $\rho(T)$  below  $T_0$ .  $\delta\rho$  (right axis) as defined in the text. The lines are guides to the eye. The vertical dashed line marks the critical pressure  $P_c = 15$  kbar.

$T_{c0}=3.9$  K, which can then be used along with the values of  $T_0(P)$  and  $T_c(P)$  to quantify the fraction of the FS that is gapped by the HO transition. The results of this analysis (labelled  $n_p$ ) are contained in Figure 3(a)—where the HO transition would appear, invoking a reasonable extrapolation, to completely gap its portion of the FS near  $P_c$  [31], thus suppressing superconductivity in the vicinity of  $P_c$  as seen. Also plotted in Figure 3(a) are extracted results for  $n(P)$  from  $C(T)$  data under pressure (labelled  $n_c$ ) [32], which are in excellent agreement with the analysis of the  $\rho(T)$  data presented herein, although  $C(T)$  data to higher pressures would be desirable to confirm this postulate.

The electrical resistivity of Figure 1(a) was fit from approximately 2 K up to 90% of  $T_0$  with the previously

employed expression [4, 33, 34]:

$$\rho(T) = \rho_0 + AT^2 + B \frac{T}{\Delta} \left(1 + \frac{T}{\Delta}\right) e^{(-\Delta/T)}, \quad (2)$$

which accounts for the residual resistivity  $\rho_0$ , a heavy Fermi liquid term, and scattering from gapped spin excitations. The magnitude of the magnon gap  $\Delta$  was found to undergo a change in its pressure dependence near  $P_c$ , as shown in Figure 3(b), where the error bars result from the fitting algorithm used. Within a SDW formalism, this magnon dispersion gap  $\Delta$  is associated with the magnitude of the FS gap [35]; and, although other conceivable pressure-dependent parameters are involved in the relationship,  $\Delta(P)$  is an indirect probe of the pressure-dependent evolution of the FS. Also shown in Figure 3(b) is the magnitude of the HO anomaly  $\delta\rho$ , defined by extrapolating the temperature dependences above and below the HO transition to  $T_0$  and evaluating the difference in the resultant electrical resistivity; the ascribed error bars result from small differences in permissible extrapolations. The quantity  $\delta\rho$  decreases with pressure up to  $P_c$ , after which it remains constant. This behavior can be understood within the context of the fraction of FS left ungapped by the HO transition: with applied pressure, the fraction of FS gapped by the HO transition grows larger and reduces the number of available states into which quasiparticles can scatter, resulting in a reduction in the scattering and a consequent reduction in  $\delta\rho$ ; at and above  $P_c$ , the FS is completely gapped and the scattering processes along with  $\delta\rho$  become constant.

The salient features of the results presented herein can be summarized as follows: (1) the HO state and its associated qualitative features persist in  $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ , suggesting that Re substitution does little to alter the HO state; (2) a distinct kink in  $T_0(P)$  is seen at  $P_c=15$  kbar and the kink is independent of the value of  $T_0(0)$  as modified by increasing  $x$ ; (3) the HO state coincides with superconductivity, and the mechanism for superconductivity appears unaltered according to the variations of  $T_c$  and  $H_{c2}(T)$  with  $P$ ; (4) the HO and SC states compete for FS fraction, with the former occupying 100% of its portion of FS near 15 kbar; (5) the gap  $\Delta$  associated with magnetic excitations inferred from  $\rho(T, P)$  measurements evinces a change in behavior near 15 kbar. It would appear likely that at the critical pressure of  $P_c=15$  kbar,  $\text{URu}_2\text{Si}_2$  undergoes a distinct HO/AFM transition, although the degree of order and nature of the transition remain uncertain. Furthermore, this HO/AFM transition seemingly occurs along a vertical or near vertical phase boundary near 15 kbar. The coincidence of the HO/AFM boundary, the fully gapped portion of the FS where HO resides, and the change in  $\Delta$  is not currently understood. No particular microscopic model has been invoked to discuss the results, although the simultaneous

presence of a spin and FS gap strongly favor the formation of a SDW-like FS instability at  $T_0$ .

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- [1] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **55**, 2727 (1985).
  - [2] W. Schlitz *et al.*, Z. Phys. B **62**, 171 (1986).
  - [3] M. B. Maple *et al.*, Phys. Rev. Lett. **56**, 185 (1986).
  - [4] M. W. McElfresh *et al.*, Phys. Rev. B **35**, 43 (1987).
  - [5] K. Behnia *et al.*, Phys. Rev. Lett. **94**, 156405 (2005).
  - [6] P. A. Sharma *et al.*, Phys. Rev. Lett. **97**, 156401 (2006).
  - [7] C. Pfleiderer, J. A. Mydosh, and M. Vojta, Phys. Rev. B **74**, 104412 (2006).
  - [8] C. Broholm *et al.*, Phys. Rev. Lett. **58**, 1467 (1987).
  - [9] P. Santini, and G. Amoretti, Phys. Rev. Lett. **73**, 1027 (1994).
  - [10] A. Kiss, and P. Fazekas, Phys. Rev. B **71**, 054415 (2005).
  - [11] Y. Okuno, and K. Miyake, J. Phys. Soc. Jpn. **67**, 2469 (1987).
  - [12] C. M. Varma and L. Zhu, Phys. Rev. Lett. **96**, 036405 (2006).
  - [13] P. Chandra *et al.*, Nature **417**, 1831 (2002).
  - [14] H. Ikeda and Y. Ohashi, Phys. Rev. Lett. **81**, 3723 (1998).
  - [15] V. P. Mineev and M. E. Zhitomirsky, Phys. Rev. B **72**, 014432 (2005).
  - [16] F. Bourdarot *et al.*, Phys. Rev. Lett. **90**, 067203 (2003).
  - [17] C. R. Wiebe *et al.*, Nature Physics **3**, 522 (2007).
  - [18] D. A. Bonn, J. D. Garrett, and T. Timusk, Phys. Rev. Lett. **61**, 1305 (1988).
  - [19] K. H. Kim *et al.*, Phys. Rev. Lett. **91**, 256401 (2003).
  - [20] N. K. Sato *et al.*, Physica B **378-380**, 576 (2006).
  - [21] H. Amitsuka *et al.*, J. Magn. Magn. Mater. **310**, 214 (2007).
  - [22] G. Knebel *et al.*, J. Magn. Magn. Mater. **310**, 195 (2007).
  - [23] H. Amitsuka *et al.*, Phys. Rev. Lett. **83**, 5114 (1999).
  - [24] F. Bourdarot *et al.*, Physica B **359-361**, 986 (2005).
  - [25] K. Matsuda *et al.*, J. Phys.: Condens. Matter **15**, 2363 (2003).
  - [26] H. Amitsuka *et al.*, Physica B **326**, 418 (2003).
  - [27] E. D. Bauer *et al.*, Phys. Rev. Lett. **94**, 046401 (2005).
  - [28] G. Bilbro and W. L. McMillan, Phys. Rev. B **14**, 1887 (1976).
  - [29] G. Motoyama, T. Nishioka, and N. K. Sato, Phys. Rev. Lett. **90**, 166402 (2003).
  - [30] V. A. Sidorov and R. A. Sadykov, J. Phys.: Condens. Matter **17**, S3005 (2005).
  - [31] The presence of metallic conduction below  $T_0$  indicates that only a portion of the entire FS is gapped by the HO transition, leaving residual density of states at the Fermi level to prohibit a metal-insulator transition, consistent with  $\text{URu}_2\text{Si}_2$  being a multiband material. See, for example, J. D. Denlinger *et al.*, J. Electron Spectrosc. Relat. Phenom. **117-118**, 347 (2001).
  - [32] R. A. Fisher *et al.*, Physica B **163**, 419 (1990).

- [33] N. Hessel Andersen, in *Crystalline Electric Field and Structural Effects in f-electron Systems*, edited by J. E. Crow, R. P. Guertin, and T. W. Mihalisin (Plenum, New York, 1980) p.373.
- [34] T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. B **33**, 6527 (1986).
- [35] G. Grüner, Rev. Mod. Phys. **66**, 1 (1994).